

Estimating Sulfuric Acid Aerosol Emissions from Coal-Fired Power Plants

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ABSTRACT

This paper presents a proposed method for estimating the release of sulfuric acid (H_2SO_4) aerosol from coal-fired power plants. The U. S. Environmental Protection Agency's Toxic Release Inventory (TRI) reporting system requires that, beginning in 1998, electric utilities must estimate their emissions of over 600 chemical compounds. Aerosols of H_2SO_4 are one of the compounds included in the TRI. In coal-fired plants, H_2SO_4 vapor is created in the ductwork downstream of the boiler by the combination of water vapor and sulfur trioxide (SO_3), both of which are produced during the coal combustion process. In some cases, the vapor condenses as an aerosol, which makes it a reportable compound. Since the TRI system does not require the collection of any new data, the method presented herein was developed to use data and information already available at most coal-fired plants. These factors include the SO_2 emission rate, the type of fuel being burned, and the particulate control device used to control dust emissions.

INTRODUCTION

In June 1995, the U. S. Environmental Protection Agency modified the list of chemicals subject to reporting requirements under Section 313 of the Emergency Planning and Community Right-to-Know Act (EPCRA) [1]. At that time, the reportable forms of sulfuric acid (H_2SO_4) were changed to include only acid aerosols. Beginning in 1998, most electric utilities are required to report emissions of these sulfuric acid aerosols if they meet any of the following annual threshold requirements [2]:

1. If the facility *manufactures* 25,000 pounds of sulfuric acid aerosols
2. If the facility *processes* 25,000 pounds of sulfuric acid aerosols
3. If the facility otherwise *uses* 10,000 pounds of sulfuric acid aerosols.

This paper presents a proposed method for estimating the release of H_2SO_4 aerosols from coal-fired utility boilers (item 1 above). In coal-fired plants, H_2SO_4 vapor forms in the ductwork downstream of the boiler by the combination of water vapor and sulfur trioxide (SO_3), both of which are produced during the coal combustion process. After formation, some of the H_2SO_4 may condense and be emitted as an aerosol (the reportable quantity) depending on the flue gas temperature.

Following the presentation of the proposed prediction method, supporting information regarding the impact of fuel grade on the formation of SO_3 , the conversion of SO_3 to an H_2SO_4 vapor, the effect of particulate control equipment on H_2SO_4 collection, and the condensation of H_2SO_4 to an acid aerosol is presented. The effect of coal sulfur content is also shown. Sample calculations are provided.

PREDICTION METHOD

The following set of relationships is proposed to predict the H_2SO_4 aerosol concentrations in the flue gas exiting a coal-fired power plant:

$$E1 = E2 - E3 \quad (1)$$

where,

$E1$ = H_2SO_4 aerosol concentration, ppm (parts per million)

$E2$ = Total H_2SO_4 concentration, ppm

$E3$ = H_2SO_4 vapor concentration, ppm

The total H_2SO_4 concentration is determined with the following relationship:

$$E2 = K \cdot F1 \cdot F2 \cdot E4 \quad (2)$$

where,

K = Molecular weight and units conversion constant = $98.07 / 64.04 = 1.53$

98.07 = Molecular weight of H_2SO_4 ; 64.04 = Molecular weight of SO_2

$F1$ = Fuel Impact Factor

$F2$ = Technology Impact Factor

$E4$ = Sulfur dioxide (SO_2) emissions concentration, ppm

The H₂SO₄ vapor fraction (E3) is based on the total H₂SO₄ concentration and the flue gas exit temperature.

In the derivation of this relationship, the following assumptions are made:

- The grade of coal being burned impacts the rate of conversion from SO₂ to SO₃.
- All SO₃ is converted to an H₂SO₄ vapor.
- The type of particulate control device at the plant impacts the flue gas concentration of H₂SO₄.
- The boiler is wall-fired or tangentially fired.
- The rate of SO₃ formation is independent of the boiler firing rate (unit load).

Supporting information and specific values for determining the SO₂ concentration (E4), the H₂SO₄ vapor fraction (E3), the Fuel Impact Factor (F1), and the Technology Impact Factor (F2) are provided in the section below. Finally, a method to determine the portion of H₂SO₄ vapor that forms an acid aerosol is provided. Sample calculations with applications of this relationship to a specific plant configuration are provided in the following section.

SUPPORTING INFORMATION

SO₂ Concentration (E4)

The concentration of SO₂ in the flue gas is determined by conversion from the annual emissions rate as shown in the relationship below [3]:

$$E4 = \frac{2000 \cdot E5}{F \cdot k_{SO_2} \cdot HI} \cdot \left(\frac{20.9 - \%O_{2,d}}{20.9} \right) \quad (3)$$

where:

- E4 = SO₂ flue gas concentration, ppm
- E5 = Annual SO₂ emissions as recorded by a continuous emissions monitor, tons per year
- F = Fuel factor
= 9780 dscf / MBtu for bituminous and sub-bituminous coals
- k_{SO₂} = Conversion factor
= 1.660 x 10⁻⁷ (lb/scf)/ppm SO₂
- HI = Annual heat input for the unit, MBtu / year
- %O_{2,d} = Concentration of oxygen at the stack (dry). A typical flue gas oxygen concentration at the stack is 6 percent.

Fuel Impact Factor (F1)

In coal-fired electric utility boilers, gaseous sulfur-bearing compounds are generated during the combustion process. The majority of the sulfur in the coal combines with oxygen to form SO₂. However, a small minority of the sulfur is further oxidized to form SO₃. The formation of SO₃ is a complex process that is not thoroughly understood even after many years of investigation [4, 5]. The rate of formation is dependent upon a number of factors such as the sulfur content of the fuel, amount of excess air, and the presence of some form of catalyst.

Three mechanisms have been proposed to explain the oxidation of SO₂ to SO₃ [6, 7].

1. Oxidation of SO₂ in the flame by atomic oxygen

$$\text{SO}_2 + \text{O} \leftrightarrow \text{SO}_3$$
2. Oxidation of SO₂ by molecular oxygen

$$\text{SO}_2 + \frac{1}{2} \text{O}_2 \leftrightarrow \text{SO}_3$$
3. Catalytic oxidation via molecular oxygen.

This last mechanism is a result of catalytic oxidation of SO₂ to SO₃ by both ash particles and metal heat transfer surfaces. This oxidation occurs at the temperature range (800 – 1100°F) found in the economizer section of utility boilers.

In the literature, varying and sometimes conflicting estimates exist regarding the conversion of SO₂ to SO₃. For example, in one publication the conversion rate is estimated to vary from 3 to 5 percent, from 1.25 to 5 percent, and from 1 to 4 percent, depending on the section of the book being read [8]. In other reports, which focus on the performance of cold-side ESPs, the ratio of SO₂ to SO₃ at the air heater outlet is presented. These ratios are lower since a portion of the SO₃ generated during the coal combustion process condenses onto the cold sections of the air heater baskets as the flue gas temperature drops. For example, in one evaluation average flue gas SO₃ concentrations dropped from 25 ppm to 11 ppm (56 percent) across a hot-side ESP and an air heater [9]. Other reports, such as an EPA-documented SO₂ to SO₃ ratio of 0.4 percent [10], confirm these pilot-scale results. The same EPA study reports that the SO₃ levels from six different power stations varied from undetectable levels to 0.67 percent of the SO₂ concentration (Table 1). Other full-scale experimental results based on measurements during 16 field tests showed concentrations ranging from 0.1 to 0.41 percent of the SO₂ levels (Table 2) [11]. In both of these examples, the SO₃ concentrations when burning western coals were lower than the SO₃ concentrations when burning eastern coals. Laboratory analyses have confirmed the directly proportional relationship between the SO₂ to SO₃ conversion rate and the sulfur content of the fuel [12].

Based on the data provided above, proposed values for the Fuel Impact Factor (F1) are provided in Table 3. These factors are based on the full-scale plant data presented previously and account for SO₃ condensation that occurs in the air heater. In proposing these factors, corroborating and / or conflicting information regarding the Fuel Impact Factor for these and other fuels is sought.

Table 1. SO₂ and SO₃ measurements from 6 different power plants

Station Number	SO ₂ , ppm	SO ₃ , ppm	SO ₃ / SO ₂ ratio
1 (western coal)	262	<1	<0.0038
5 (western coal)	480	<1	<0.0021
13 (western coal)	430	<1	<0.0023
3 (eastern coal)	2440	6-9	0.0025 – 0.0037
4 (eastern coal)	755	2-3	0.0026 – 0.0040
7 (eastern coal)	600	3-4	0.0050 – 0.0067

Table 2. Ratio of SO₃ / SO₂ based on in situ measurements for 16 field tests

Coals Burned	SO ₃ / SO ₂ ratio	
	average	standard deviation
9 eastern bituminous coals	0.0041	0.0027
7 western subbituminous and lignite coals	0.0011	0.0005

Table 3. Fuel Impact Factors for various coals

Coal	F1
Eastern bituminous	0.004
Western bituminous	0.001
Powder River Basin (PRB)	0.0005

Technology Impact Factor (F2)

Electrostatic precipitators and baghouses remove particulates from the flue gas stream. In the process, they also remove acid gases that have condensed onto the particulate matter. Hot-side ESPs function at temperatures where little H₂SO₄ has formed. However, in cold-side ESPs and baghouses, a majority of the SO₃ has converted to H₂SO₄ vapor. As the flue gas temperature is reduced, H₂SO₄ aerosols adsorb onto the fly ash to form an acid layer that allows electrical currents to flow more easily, thereby lowering the ash resistivity. The amount of H₂SO₄ interacting with the fly ash increases as the temperature decreases [13].

Little definitive data are available describing the SO₃ / H₂SO₄ collection rate in cold-side particulate collection equipment. However, anecdotal evidence suggests that acid aerosol collection does occur. For example, in flue gas from the combustion of low-sulfur coal (low native SO₃ concentrations), SO₃ injection systems are used for flue gas treatment to improve particulate collection efficiency. The SO₃ injection rate is controlled to provide optimal performance with minimal SO₃ utilization. In doing this, SO₃ injection is increased to the point where no appreciable increases in ESP performance occur (i.e., no additional H₂SO₄ is being adsorbed by the ash). With respect to baghouses, the dust cake on the bags acts as a barrier filter through which all H₂SO₄ vapor must pass. It is assumed that a majority of the H₂SO₄ condenses and is adsorbed by the fly ash as it passes through the dust cake.

Technology Impact Factors (F2) for ESPs and baghouses are proposed (Table 4). In sites where multiple devices are installed, the factor for the device nearest the stack should be used. These factors are engineering estimates based on available data and operational experience. In proposing these factors, corroborating and / or conflicting information is sought.

Table 4. Technology Impact Factors for particulate control devices

Particulate Control Device	F2
Hot-side ESP	1.00
Cold-side ESP	0.75
Baghouse	0.10

Sulfuric Acid (H₂SO₄) Vapor Formation

This analysis assumes that all of the SO₃ forms an H₂SO₄ vapor prior to reaching the stack exit. Sulfur trioxide is a hygroscopic material and will absorb moisture at temperatures well above its dewpoint [14, 15, 16]. By definition, a hygroscopic material has solvent properties, and its moisture content will approach equilibrium with the moisture content of the surrounding air. As the flue gas temperature drops, the concentration of H₂SO₄ increases and the concentration of SO₃ decreases. For example, in 400°F flue gas with a moisture content of 8 percent, nearly 99 percent of the SO₃ has combined with water to form an H₂SO₄ vapor (Table 5, Figure 1) [17]:

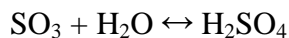


Table 5. SO₃ conversion to H₂SO₄ vapor at various flue gas temperatures

Temperature, °F	SO ₃ converted to H ₂ SO ₄ , %
800	3.85
700	14.30
600	47.54
550	70.54
500	87.50
400	98.86
350	99.74

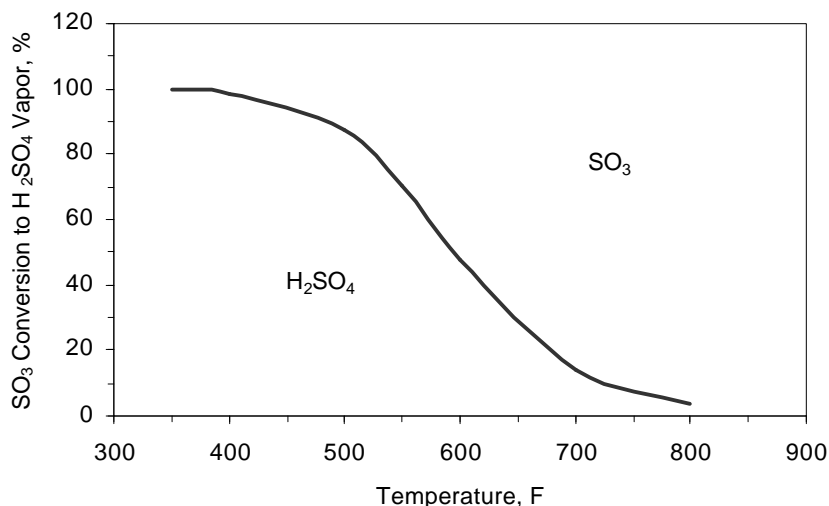


Figure 1. Portion of SO₃ that is converted to H₂SO₄ vapor at various flue gas temperatures assuming flue gas moisture content of 8 percent.

H₂SO₄ Vapor Fraction (E3)

The portion of H₂SO₄ that condenses from the vapor to an aerosol (the reportable quantity) is a function of the total H₂SO₄ concentration, the flue gas temperature, and the flue gas moisture content. In cases where the stack flue gas exit temperature exceeds the H₂SO₄ dew point, no acid aerosols are formed. In other words, the total H₂SO₄ concentration, E2, is equal to the H₂SO₄ vapor concentration, E3, and the reportable quantity is zero.

However, when the stack flue gas exit temperature is at or below the H₂SO₄ dew point, an acid aerosol / vapor mixture is present. As a result, the vapor fraction must be calculated to determine the reportable acid aerosol concentration. To accomplish this, the H₂SO₄ dew point must be calculated.

The H₂SO₄ dew point is a function of the partial pressures of the H₂SO₄ and the moisture in the flue gas. It can be approximated by the following relationship [18]:

$$1/DP = 2.276 \times 10^{-3} - 2.943 \times 10^{-5} \ln(pp_{H_2O}) - 8.58 \times 10^{-5} \ln(pp_{H_2SO_4}) + 6.20 \times 10^{-6} \ln(pp_{H_2O}) \ln(pp_{H_2SO_4}) \quad (4)$$

where:

- DP = H₂SO₄ dew point, °Kelvin
- pp_{H₂O} = partial pressure of the flue gas moisture, mm Hg
= 760 · (flue gas moisture content, %)
- pp_{H₂SO₄} = partial pressure of the H₂SO₄, mm Hg
= 760 · (H₂SO₄ concentration, ppm) · 1 × 10⁻⁶

This relationship is shown graphically for flue gas with a moisture content of 10 percent in Figure 2. Above the curve, all H₂SO₄ exists as a vapor. Below the curve, an aerosol/vapor mixture is present. Dew points for various H₂SO₄ concentrations at two different flue gas moisture contents are provided in Table 6.

Using Table 6 or Figure 3, the vapor fraction (E3) for a specific flue gas temperature can be determined. This vapor fraction also can be used to determine the H₂SO₄ aerosol fraction in the flue gas. An example graphical determination of the vapor fraction and the aerosol fraction is provided in Figure 3. In a flue gas determined to have a 9-ppm H₂SO₄ concentration (E2), a moisture content of 10 percent, and a stack exit gas temperature of 250°F, the vapor fraction (E3) is approximately 1.7 ppm. The aerosol fraction would be approximately 7.3 ppm.

Table 6. H₂SO₄ dew points (°F) for two different flue gas moisture contents

H ₂ SO ₄ vapor concentration, ppm	8% moisture	10% moisture
0.1	200.6	205.4
0.5	224.9	229.5
1	236.0	240.5
3	254.3	258.5
5	263.1	267.3
10	275.4	279.5
15	282.9	286.8
20	288.2	292.1

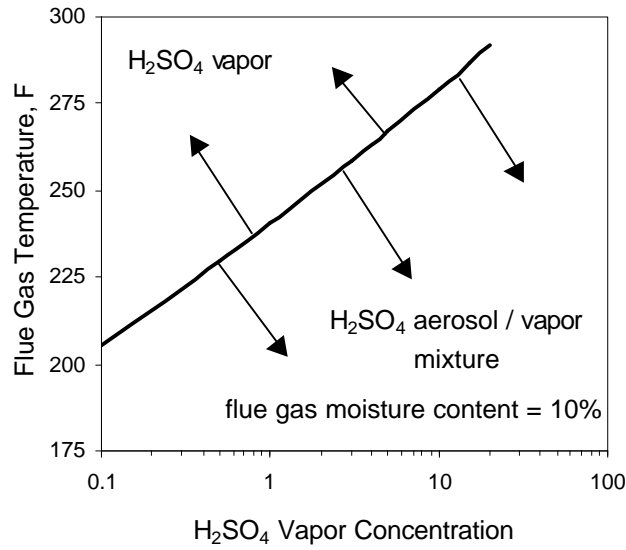


Figure 2. H_2SO_4 aerosol formation diagram (log-linear scale).

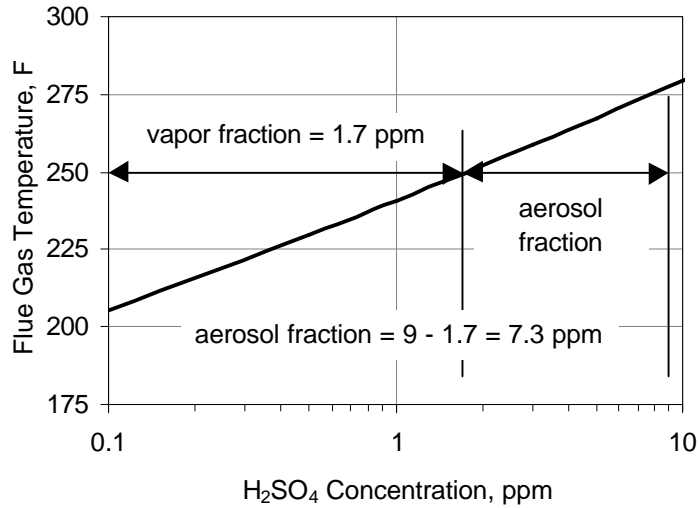


Figure 3. Relationship between vapor fraction and aerosol fraction (log-linear scale).

H_2SO_4 Emission Rate

Once the H_2SO_4 aerosol concentration (ppm) is determined, the H_2SO_4 emissions rate (pounds per year) can be calculated using the following relationship [19]:

$$E6 = E1 \cdot k_{\text{H}_2\text{SO}_4} \cdot F \cdot \text{HI} \cdot \left(\frac{20.9}{20.9 - \% \text{O}_{2,d}} \right) \quad (5)$$

where:

$$\begin{aligned} E6 &= \text{H}_2\text{SO}_4 \text{ aerosol emissions rate, pounds per year} \\ k_{\text{H}_2\text{SO}_4} &= \text{Conversion factor} = K \cdot k_{\text{SO}_2} \\ &= 2.54 \times 10^{-7} \text{ (lb/scf)/ppm H}_2\text{SO}_4 \end{aligned}$$

EXAMPLE CALCULATIONS

This section presents a series of example calculations based on the solution process provided above.

Example 1. A base-loaded, 700-MW coal-fired boiler equipped with a cold-side ESP burns an eastern bituminous coal. SO₂ emissions are 33,700 tons per year. Annual heat input is 43,800,000 MBtu. Flue gas moisture content is 10 percent and stack oxygen concentration is 6 percent (dry). Based on data from continuous emissions monitor RATA tests, the stack exit gas temperature at full load is 297°F.

Solution 1. Determine flue gas SO₂ concentration (Equation 3).

$$E4 = \frac{2000 \cdot 33,700}{9780 \cdot 1.660 \times 10^{-7} \cdot 43,800,000} \cdot \left(\frac{20.9 - 6}{20.9} \right) = 675 \text{ ppm}$$

Determine total H₂SO₄ concentration in the flue gas at the stack (Equation 2).

$$E2 = 1.53 \cdot 0.004 \cdot 0.75 \cdot 675 = 3.1 \text{ ppm}$$

Using Table 6, the dew point for an H₂SO₄ concentration of 3.1 ppm is approximately 260°F. Since, the stack exit temperature exceeds the dew point, H₂SO₄ aerosol emissions are zero.

Example 2. Use the same information from example 1; however, in this case unit operation cycles. Stack exit gas conditions and unit operations data for three different load points are available (below).

Condition	Stack Temperature, °F	Heat Input, MBtu
Full load (700 MW)	297	30,000,000
Mid load (500 MW)	249	7,000,000
Low load (340 MW)	229	6,800,000

Solution 2. From Example 1, the total H₂SO₄ concentration in the flue gas at the stack is 3.1 ppm. Now, consider H₂SO₄ the vapor / aerosol fractions for each operating condition.

Full-load operation. From Example 1, all H₂SO₄ emissions are in the vapor state at 297°F. There are no aerosol H₂SO₄ emissions.

Mid-load operation. Using Table 6, the mid-load stack exit gas temperature is below the dew point for an H₂SO₄ concentration of 3.1 ppm. Some aerosols will be

present. By interpolation in Table 6, the vapor concentration is approximately 2 ppm at 249°F. Applying Equation 1, the H₂SO₄ aerosol concentration is 3.1 – 2.0 = 1.1 ppm. Applying Equation 5, the H₂SO₄ aerosol emissions rate for this load bin can be determined:

$$E6 = 1.1 \cdot 2.54 \times 10^{-7} \cdot 9780 \cdot 7,000,000 \cdot \left(\frac{20.9}{20.9 - 6.0} \right) = 24,400 \text{ pounds per year.}$$

Low-load operation. Using Table 6, the low-load stack exit gas temperature is below the dew point for an H₂SO₄ concentration of 3.1 ppm. Some aerosols will be present. By interpolation in Table 6, the vapor concentration is approximately 0.7 ppm at 229°F. Applying Equation 1, the H₂SO₄ aerosol concentration is 3.1 – 0.7 = 2.4 ppm. Applying Equation 5, the H₂SO₄ aerosol emissions for this load bin can be determined:

$$E6 = 2.4 \cdot 2.54 \times 10^{-7} \cdot 9780 \cdot 6,800,000 \cdot \left(\frac{20.9}{20.9 - 6.0} \right) = 56,900 \text{ pounds per year.}$$

The total H₂SO₄ aerosol emission is the sum of the emissions from the three load bins: 0 + 24,400 + 56,900 = 81,300 pounds per year.

OTHER IMPACTS

In addition to the factors considered above, the following site-specific characteristics may impact H₂SO₄ emissions rates. These factors are not considered in this proposed method.

1. NO_x reduction catalysts. Typically, catalysts used to reduce NO_x emissions increase flue gas SO₃ concentrations. Results from a DOE-sponsored evaluation of several different catalysts treating flue gas from the combustion of high-sulfur U.S. coals reported average SO₃ oxidation levels of 0.31 percent [20].
2. Ammonia injection systems. Ammonia injection systems are used to improve the performance of cold-side ESPs and in NO_x control technologies (selective catalytic reduction and selective non-catalytic reduction.) At temperatures below approximately 500°F, ammonia preferentially reacts with SO₃ to form ammonium sulfate and ammonium bisulfate. These reactions can act to reduce or eliminate the presence of SO₃ (and subsequently H₂SO₄) in the flue gas stream.
3. SO₃ injection systems. SO₃ injection systems (sometimes called sulfur burners) are used to improve the particulate collection capabilities of cold-side ESPs. The SO₃ is injected into the flue gas upstream of the ESP. The SO₃ reacts with moisture in the flue gas to create H₂SO₄, which improves the resistivity of the fly ash making it easier to collect. Some of the H₂SO₄ (~1 ppm) generated by the sulfur burners may slip through the ESP resulting in higher flue gas H₂SO₄ concentrations at the stack.
4. Natural gas co-firing. Since natural gas does not contain sulfur, co-firing or reburning natural gas in a boiler may change the SO₃ to SO₂ production ratio.

CONCLUSIONS

This paper presents a method for predicting the emissions of sulfuric acid aerosols from coal-fired power plants. The calculation is based on the unit's SO₂ emission rate as recorded by a continuous emissions monitor. The method accounts for the rank of coal being burned and the

type of particulate control device installed at the facility. The effects of H₂SO₄ vapor condensation at the dew point and subsequent aerosol formation are included in the analysis. Influencing factors not accounted for in the process include catalytic conversion across NO_x reducing catalysts, reactions with ammonia injected for NO_x control or flue gas conditioning, additional SO₃ injected for flue gas conditioning, and natural gas co-firing.

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